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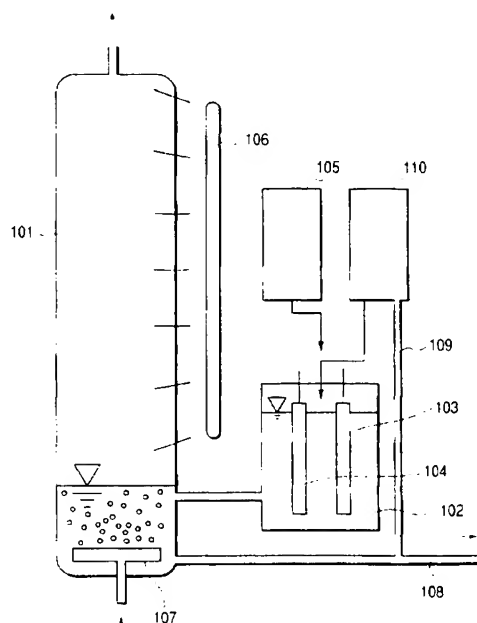
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(54) **Process and system for decomposing pollutants**

(57) In a pollutant decomposition process for decomposing pollutants by bringing pollutants contained in air into contact with air which contains chlorine, under irradiation by light, at least part of a chlorine-generating

solution present in a chlorine generation region is fed to means for forming functional water by electrolysis to effect regeneration and is again fed to the chlorine generation region. Also disclosed is a pollutant decomposition system used in such a process.

FIG. 1



Description

BACKGROUND OF THE INVENTION

Field of the Invention

[0001] This invention relates to a process for decomposing pollutants (in particular, organochlorine compounds) and a pollutant decomposition system used therefor.

Related Background Art

[0002] With development of industrial techniques until recent years, organochlorine compounds such as ethylene chloride and methane chloride have enormously been put into use, and their disposal has come into serious question. An environmental problem that these pollutants derived after use pollute natural environment has also been brought out, and great efforts are being made for its solution.

[0003] As methods for disposing of such pollutants, for example, methods are available in which ethylene chloride is decomposed with an oxidant or a catalyst. Stated specifically, known are a method in which it is decomposed with ozone (Japanese Patent Application Laid-Open No. 3-38297) and a method in which it is irradiated by ultraviolet rays in the presence of hydrogen peroxide (Japanese Patent Application Laid-Open No. 63-218293). It is also suggested to use sodium hypochlorite as an oxidizing agent (U.S. Patents No. 5,525,008 and No. 5,611,642). Also proposed is a method in which sodium hypochlorite and ultraviolet irradiation are used in combination (U.S. Patent No. 5,582,741). Another method is also known in which a photocatalyst comprised of fine semiconductor particles of an oxide such as titanium oxide and liquid ethylene chloride are suspended under an alkaline condition, and the suspension is irradiated by light to effect decomposition (Japanese Patent Application Laid-Open No. 7-144137).

[0004] In addition to the foregoing, methods of photodecomposition made by ultraviolet irradiation in a gaseous phase without use of any oxidizing agent have already been attempted. For example, proposed are a method in which waste gas containing organohalogen compounds is subjected to ultraviolet irradiation to convert it into an acidic decomposed gas, followed by washing with an alkali to make it harmless (Japanese Patent Application Laid-Open No. 62-191025), and a system in which waste water containing organohalogen compounds is subjected to aeration and the gas being discharged is subjected to ultraviolet irradiation, followed by washing with an alkali (Japanese Patent Application Laid-Open No. 62-191095). It is also known to decompose ethylene chloride by iron powder (Japanese Patent Application Laid-Open No. 8-257570). In this case, it is presumed that reduction decomposition takes place.

Reduction decomposition is also reported in respect of decomposition of tetrachloroethylene (hereinafter abbreviated to "PCE") by the use of fine silicon particles.

[0005] Chlorinated aliphatic hydrocarbons such as trichloroethylene (hereinafter abbreviated to "TCE") and PCE are known to be aerobically or anaerobically decomposed by microorganisms, and it is also attempted to make decomposition or purification by utilizing such a process.

SUMMARY OF THE INVENTION

[0006] An object of the present invention is to provide a decomposition process which does not require any treatment with activated carbon or microorganisms and by which pollutants can be decomposed efficiently and without causing any secondary pollution on account of the use of air which contains chlorine, and also which may produce waste water in a small quantity; and an efficient pollutant decomposition system employing such a process.

[0007] To achieve the above object, the present inventors made extensive studies. As a result, they have reached a new finding that superior decomposition power can be achieved by subjecting functional water (e.g., acidic water) to aeration to form air which contains chlorine, and mixing this air with air which contains pollutants such as organochlorine compounds, followed by photodecomposition; the functional water being obtained by electrolysis of water that is reported to have a microbicidal effect (Japanese Patent Application Laid-Open No. 1-180293) or the effect of cleaning contaminants present on semiconductor wafers (Japanese Patent Application Laid-Open No. 7-51675).

[0008] In the course of detailed experiments continued, further making additional studies thereafter on any practically desirable form, they have also discovered that, in order to materialize simpler and more efficient decomposition, it is effective to carry out electrolysis on a functional-water waste liquor formed in the course of aeration or after the aeration, to form functional water again usable as a chlorine feed source so that this can be reused to carry out the decomposition, making it possible to greatly cut down the quantity of waste water and that of the electrolyte to be added. Thus, they have accomplished the present invention.

[0009] More specifically, the present invention provides a pollutant decomposition process for decomposing pollutants by bringing pollutants contained in air into contact with air which contains chlorine, under irradiation by light, the process comprising:

a chlorine-containing air generation step of generating air which contains chlorine, by bringing air into contact with a chlorine-generating solution comprised of functional water (I) or functional water (II) having been fed into a chlorine generation region; a decomposition step of decomposing the pollut-

ants by bringing the air which contains chlorine and air which contains pollutants into contact with each other under irradiation by light in a decomposition treatment region;

a regeneration step of obtaining functional water (II) by regeneration by feeding as functional water waste liquor at least part of the chlorine-generating solution present in the chlorine generation region, to means for forming functional water by electrolysis; and

a feed step of feeding to the chlorine generation region the functional water (II) obtained through the regeneration step;

the functional water (I) and functional water (II) being water capable of generating by aeration the air which contains chlorine, and the functional water (I) comprising a solution used for its formation which does not contain the functional water waste liquor and the functional water (II) comprising a solution used for its formation which contains the functional-water waste liquor at least in part.

[0010] The present invention also provides a pollutant decomposition system for decomposing pollutants by bringing pollutants contained in air into contact with air which contains chlorine, under irradiation by light, the system comprising:

a chlorine generation region into which a chlorine-generating solution comprising functional water (I) or functional water (II) is fed to bring it into contact with air to generate air which contains chlorine;

a decomposition treatment region into which the air which contains chlorine and air which contains pollutants are fed to bring them into contact with each other under irradiation by light to decompose the pollutants;

means for effecting irradiation by light;

means for forming functional water by electrolysis; and

means for feeding at least part of the chlorine-generating solution to the means for forming functional water;

the functional water (I) and functional water (II) being water capable of generating by aeration the air which contains chlorine, and the functional water (I) comprising a solution used for its formation which does not contain the functional water waste liquor and the functional water (II) comprising a solution used for its formation which contains the functional water waste liquor at least in part.

[0011] The contact of the chlorine-generating solution with the air in the chlorine-containing air generation step may be the step of sending air to the surface of the chlorine-generating solution. In order to improve efficiency, it is preferable to use the step of enlarging the area of gas-liquid contact. To enlarge the area of gas-liquid con-

tact, preferably usable is the step of jetting the chlorine-generating solution into air or subjecting the chlorine-generating solution to aeration.

[0012] According to the present invention, a pollutant decomposition process and a pollutant decomposition system used therefor can be provided by which pollutants such as organochlorine compounds can be decomposed efficiently, safely and simply in the gaseous phase under normal temperature and normal pressure, and the quantity of the electrolyte to be added and the quantity of waste water can be cut down.

BRIEF DESCRIPTION OF THE DRAWINGS

[0013]

Fig. 1 is a schematic illustration for describing the basic construction of a first embodiment of the present invention.

Fig. 2 is a schematic illustration for describing an example in which the basic construction of the first embodiment has been modified.

Fig. 3 is a schematic illustration for describing the basic construction of a second embodiment of the present invention.

DESCRIPTION OF THE PREFERRED EMBODIMENTS

[0014] Embodiments of the present invention are described below with reference to the accompanying drawings.

(Embodiment 1)

[0015] Fig. 1 shows the basic construction of an embodiment of the pollutant decomposition system of the present invention.

[0016] In Fig. 1, reference numeral 102 denotes an electrolytic cell serving as a functional-water formation means, which is internally provided with the cathode 103 and the anode 104. Reference numeral 101 denotes a decomposition treatment tank having a chlorine generation region which has an air diffusion means 107 for aerating functional water kept at the bottom, and its decomposition treatment region is irradiated by light from a light irradiation means 106.

[0017] First, untreated water held fully in the electrolytic cell 102 is mixed with a high-concentration electrolyte solution fed from an electrolyte solution feed unit 105 to come into an aqueous electrolyte solution having a stated concentration. Not shown particularly in the drawing, a stirrer may be provided in the electrolytic cell 102, which is preferable because an aqueous electrolyte solution having a uniform concentration can be prepared in a short time by stirring the untreated water. In this state, the cathode 103 and the anode 104 are connected to a direct-current power unit (not shown) to carry

out electrolysis for a certain time to obtain functional water (I). This water is supplied to the decomposition treatment tank 101 at one time in its entirety in the case of batch operation, or at a constant flow rate in the case of continuous operation.

[0018] The functional water (I) may also be prepared without relying on electrolysis and by adding hypochlorous acid or the like. In such a case, the necessary reagent may be added to the untreated water held fully in the electrolytic cell 102 to form functional water (I), which is then supplied to the decomposition treatment tank 101 at one time in its entirety in the case of batch operation, or at a constant flow rate in the case of continuous operation. In the case of batch operation, the untreated water may directly be supplied to the bottom of the decomposition treatment tank 101 through a water supply means (not shown) and then the necessary reagent may be added to form the functional water (I).

[0019] The functional water (I) supplied to the decomposition treatment tank 101 is aerated by the air diffusion means (aeration means) 107 provided in the chlorine generation region at the bottom portion of the decomposition treatment tank 101, so that the interior of the decomposition treatment tank 101 is filled with air which contains chlorine and the decomposition treatment region is formed. Here, the air supplied to the aeration means 107 may be air which does not contain any pollutants and air which contains pollutants may separately be supplied to the decomposition treatment region of the decomposition treatment tank 101. In such a case, the air which contains pollutants may be supplied to the aeration means 107 so that mixed air of the air which contains chlorine and the air which contains pollutants may be made up in the decomposition treatment region in the decomposition treatment tank 101. This makes construction simple to some extent. Then, this mixed air may be irradiated by light from the light irradiation means 106 for a desired residence time, whereby the decomposition target substance is decomposed.

[0020] Functional water waste liquor having decreased in the amount of dissolved chlorine as a result of the aeration in the chlorine generation region inside the decomposition treatment tank 101 in the course of or after the desired decomposition reaction is discharged out of the chlorine generation region of the decomposition treatment tank 101 through a waste liquor pipe 108 at one time in its entirety in the case of batch operation, or at a constant flow rate in the case of continuous operation. Then, part or the whole of this functional water waste liquor is flowed back to the electrolytic cell 102 through a functional-water waste liquor flow-back pipe 109. Also, a storage tank 110 may optionally be provided in the course of the functional water waste liquor flow-back pipe 109 so that the functional water waste liquor can temporarily be stored.

[0021] In the case where it is not flowed back in its entirety and is partly flowed out, the untreated water must anew be added to the electrolytic cell 102 in a

quantity corresponding to that of flow-out.

[0022] The functional water waste liquor in this embodiment may come to have a pH close to 4, depending on the pH of the original functional water and the feed of pollutants. This waste liquor may be flowed back, as it is, to the decomposition treatment tank 101 and electrolyzed to make up functional water (II), or may be neutralized and thereafter electrolyzed. In the case where the waste liquor is drained, it should be subjected to neutralization. As an aqueous alkaline solution used for such neutralization, an aqueous solution of an alkali reagent such as sodium hydroxide may be used. Not shown in the drawing, a means for mixing the alkali reagent in the functional water waste liquor while monitoring the pH may also additionally be provided in the course of the waste liquor pipe 108.

[0023] The functional water waste liquor flowed back to the electrolytic cell 102 is electrolyzed there, and comes into functional water (II) and is regenerated as a source from which the air which contains chlorine is fed.

[0024] When the functional water (II) is made up, the functional water waste liquor may preferably be regulated in some cases to have a suitable electrolyte concentration. If the electrolyte concentration has already been in a proper range, the step of such regulation may be omitted. A means may also be provided for feeding the electrolyte automatically while measuring the concentration of dissolved chlorine in the functional water present in the chlorine generation region at the bottom portion of the decomposition treatment tank 101 or measuring the concentration of electrolyte in the electrolytic cell 102 or storage tank 110. However, according to experiments made by the present inventors, it has been found that the decomposition power of the whole system does not lower when the functional water waste liquor is flowed back and regenerated in a quantity about five times the quantity of the functional water in the system and is thereafter again added. Hence, it is possible to operate the system even if any means for regulating the electrolyte concentration is not provided.

[0025] Then, the functional water waste liquor fed into this electrolytic cell 102 is electrolyzed to form functional water (II) which can again be used for the decomposition. This functional water (II) is further fed into the chlorine generation region of the decomposition treatment tank 101, and is aerated according to the same procedure as the case of the above functional water (I) to generate the air which contains chlorine, where the mixed air thereof with the pollutants is again made up to carry out the decomposition under irradiation by light in the decomposition treatment region.

[0026] Subsequently, such steps of forming the functional water (II) in the electrolytic cell 102 and decomposing the pollutants in the decomposition treatment region inside the decomposition treatment tank 101 are repeated any desired times. This makes it possible to greatly cut down the total quantity of waste water and the quantity of the electrolyte to be added.

[0027] Fig. 2 shows a decomposition system partly modified from the system shown in Fig. 1. As shown in Fig. 2, the system may be so constructed that the chlorine generation region which is the part where the functional water present at the bottom of the decomposition treatment tank 101 shown in Fig. 1 is aerated is made independent as a functional water aeration tank 201 and the mixed air of the air which contains chlorine and the air which contains pollutants, formed here, is sent to the decomposition treatment tank consisting of only the decomposition treatment region.

[0028] Not shown in the drawing, the system may also be so constructed that the air which contains pollutants is directly sent to any of the above two-type decomposition treatment tanks and air which does not contain any pollutants is sent to the aeration means in the chlorine generation region to generate the air which contains chlorine, where the mixed air of the both is made up to carry out decomposition under irradiation by light.

(Embodiment 2)

[0029] Fig. 3 shows the basic construction of a second embodiment of the pollutant decomposition system of the present invention.

[0030] The system shown in Fig. 3 differs from the one shown in Fig. 1 in that the electrolytic cell 102 is provided therein with a diaphragm 111 and that the cathode 103 side in the electrolytic cell 102 communicate with the waste liquor pipe 108 through a pipe (alkaline-water pipe) 112 at the latter's part on the side upstream to the part where the waste liquor pipe 108 and the functional-water waste liquor flow-back pipe 109 are joined. As the diaphragm 111, preferably usable is, e.g., an ion-exchange membrane.

[0031] In the case of the construction as shown in Embodiment 2, the maintenance must be made for the diaphragm, and also the system have a complicated construction. However, because of such construction, the acidic water formed in the vicinity of the anode 104 can be prevented from being mixed with the alkaline water formed in the vicinity of the cathode 103, so that functional water can be obtained which has a higher concentration of dissolved chlorine and is capable of generating chlorine gas in a large quantity.

[0032] The functional water waste liquor in the present Embodiment may also come to have a pH close to 1, depending on the feed of pollutants. This waste liquor may be flowed back, as it is, to the electrolytic cell 102 and electrolyzed to make up functional water (H), or may be neutralized and thereafter electrolyzed. In the case where the waste liquor is drained, it should be subjected to neutralization. As an aqueous alkaline solution used for such neutralization, an aqueous solution of an alkali reagent such as sodium hydroxide may be used. The use of alkaline water formed on the cathode side when acidic water is made up is preferred because it is unnecessary to use any alkali agent additionally or use

any unit for feeding it. This alkaline water may also be supplied through the alkaline-water pipe 112 to the functional water waste liquor present in the waste liquor pipe 108 so as to be utilized for the neutralization.

[0033] In the case of Embodiment 2 also, the basic construction and procedure for the decomposition are the same as those in Embodiment 1. Also, in the case of Embodiment 2, like Embodiment 1, the system may be so constructed that the decomposition treatment tank and the functional water aeration tank are separately provided and the functional water may be aerated with the air which does not contain any pollutants, to generate the air which contains chlorine.

[0034] These steps may be repeated batch-wise any desired times, or may be carried out continuously.

[0035] In both Embodiments, the decomposition target substance is air which contains pollutants, having been vacuum-extracted from polluted soil, or air which contains pollutants, obtained by aeration of underground water having been pumped up. Accordingly, the system may also be so constructed that hydrochloric acid, sodium chloride and sodium hypochlorite are added to pollutant-dissolved water such as underground water having been pumped up from polluted soil, to make up the functional water (I), which is then subjected to the aeration to make up the mixed air of pollutants and chlorine to carry out the decomposition under irradiation by light.

[0036] Not shown in the drawing, the waste water may also be irradiated by light to effect decomposition when in both Embodiments the waste water is mixed with the pollutants in a concentration higher than the standard for waste water.

(Pollutants to Be Treated)

[0037] Pollutants to be treated may include organochlorine compounds such as chloroethylene, 1,1-dichloroethylene, cis-1,2-dichloroethylene, trans-1,2-dichloroethylene, trichloroethylene, tetrachloroethylene, chloromethane, dichloromethane and trichloromethane.

(Untreated Water Serving as Source for Functional Water)

[0038] The untreated water may be any water as long as any substance which may adsorb chlorine gas does not stand included or any substance which may react with chlorine gas without irradiation by light does not stand dissolved. Where polluted underground water is purified, the underground water itself may be used as the untreated water so that the quantity of waste water can further be cut down. Since, however, there is a problem that the pollutants having dissolved therein may evaporate as a result of the rise of water temperature at the time of electrolysis to contaminate the air surrounding the system, it is preferable to add sodium hypochlorite or the like without relying on the electrolysis.

(Functional Water (I) and (II), and Air Which Contains Chlorine, Formed therefrom)

[0039] With regard to the mixing proportion of gaseous pollutants and the air which contains chlorine, in the decomposition treatment tank, it may preferably be so regulated that the chlorine in the air is in a concentration of from 5 ppm to 1,000 ppm. Especially when the chlorine in the air is in a concentration of from 20 ppm to 500 ppm, and further from 80 ppm to 300 ppm, which may differ depending on the concentration of the substance to be treated, the substance to be treated can be decomposed in an especially remarkable efficiency.

[0040] In the present invention, the functional water is brought into contact with the air to generate the air which contains chlorine that is necessary for the decomposition. The part where the functional water is subjected to aeration which is one of preferred methods of contact has the function to feed the chlorine basically necessary for the decomposition. The gaseous-phase reaction which takes place subsequently in the decomposition treatment tank is the principal site of decomposition reaction. Hence, in the case where the generation of chlorine and the decomposition reaction are unified as shown in Fig. 1 or 3, the proportion of the gaseous-phase portion to the liquid-phase portion has a great influence on the decomposition power. More specifically, the chlorine that can be fed increases in quantity with an increase in the volume of the functional water, but the gaseous-phase portion decreases and the reaction zone of decomposition decreases. Conversely, the site of reaction increases with an increase in the gaseous-phase portion and the decomposition reaction proceeds quickly, but the feed of chlorine decreases because of a decrease in the liquid-phase portion. There are various factors such as the rate of aeration and the speed of feed of functional water. In the case where the formation of the air which contains chlorine and the region of decomposition reaction region (reaction region) are unified as shown in Fig. 1 or 3, the liquid-phase portion in the treatment tank may be in a proportion of from 5% to 30%, and preferably from 10% to 20%. Also in the case where they are not unified, as shown in Fig. 2, the proportion of the volume of the tank in which the air which contains chlorine is formed to the volume of the tank in which the decomposition reaction is carried out may also preferably be from 1:2 to 1:9 in approximation.

[0041] Here, the functional water (I) and (II) serving as the source from which the air which contains chlorine is fed refer to, e.g., water having properties such that its hydrogen ions are in a concentration (pH value) of from 1 to 4, and preferably from 2 to 3, and dissolved chlorine is in a concentration of from 5 mg/L to 150 mg/L, and preferably from 30 mg/L to 120 mg/L.

[0042] Such functional water, in particular, the functional water (II), which is regenerated functional water, can be obtained by dissolving an electrolyte such as sodium chloride or potassium chloride in the untreated wa-

ter and electrolyzing this water in a water tank having a pair of electrodes, being obtained in the vicinity of the anode thereof. Here, the electrolyte in the untreated water before electrolysis may preferably be in a concentration of, in the case of, e.g., sodium chloride, from 20 mg/L to 2,000 mg/L, and more preferably from 200 mg/L to 1,000 mg/L.

[0043] Here, in the case where the diaphragm is provided between a pair of electrodes, the acidic water formed in the vicinity of the anode can be prevented from being mixed with the alkaline water formed in the vicinity of the cathode.

[0044] As the diaphragm, preferably usable is, e.g., an ion-exchange membrane. Then, as a means for obtaining such functional water, any commercially available generator for strongly acidic electrolytic water may be used, as exemplified by OASIS BIOHALF (trade name; manufactured by Asahi Glass Engineering Co., Ltd.) and Strong Electrolytic Water Generator Model FW-200 (trade name; manufactured by Amano K.K.).

[0045] Functional water formed from a system having no diaphragm may also be used as the functional water having been described above. For example, it is functional water having the dissolved chlorine in a concentration of from 2 mg/L to 100 mg/L, and preferably from 20 mg/L to 80 mg/L, and having a pH of from 4 to 10, and preferably from 5 to 8.

[0046] The functional water having the above properties may be not only obtained by electrolysis but also prepared by dissolving various reagents in the untreated water. For example, it may be prepared by dissolving 0.001 mol/L to 0.1 mol/L of hydrochloric acid, 0.005 mol/L to 0.02 mol/L of sodium chloride and 0.0001 mol/L to 0.01 mol/L of sodium hypochlorite. The functional water thus prepared is used as functional water put previously in the decomposition treatment tank as the functional water (I) at the time of the start of decomposition, or used when the underground water which contains pollutants is made into functional water and supplied to the decomposition treatment tank.

[0047] Functional water having a pH of 4 or above may also be not only obtained by electrolysis but also prepared by dissolving various reagents in the untreated water. For example, it may be prepared by dissolving 0.001 mol/L to 0.1 mol/L of hydrochloric acid, 0.001 mol/L to 0.1 mol/L of sodium hydroxide and 0.0001 mol/L to 0.01 mol/L of sodium hypochlorite. Alternatively, it may also be prepared by dissolving only a hypochlorite, e.g., 0.0001 mol/L to 0.01 mol/L of sodium hypochlorite. Functional water having a pH of 4.0 or below and having the dissolved chlorine in a concentration of from 2 mg/L to 2,000 mg/L may also be prepared using the hydrochloric acid and hypochlorite.

[0048] In place of the hydrochloric acid, other inorganic acid or organic acid may be used. As the inorganic acid, usable are, e.g., hydrofluoric acid, sulfuric acid, phosphoric acid and boric acid; and as the organic acid, e.g., acetic acid, formic acid, malic acid, citric acid and

oxalic acid. The functional water may also be produced using, e.g., $\text{N}_3\text{C}_3\text{O}_3\text{NaCl}_2$ commercially available as a weak acidic water generating powder (e.g., trade name: Kino-san 21X; available from Clean Chemical K.K. The functional water prepared using such chemicals also has the ability to decompose organochlorine compounds under irradiation by light like the case of the functional water obtained by electrolysis, though having a difference in decomposition power as is apparent from Examples. Here, the untreated water may include city water, river water and sea water. These types of water usually have the pH in the range of from 6 to 8 and has the dissolved chlorine in a concentration less than 1 mg/L even at maximum. Such untreated water does not have the above ability to decompose pollutants as a matter of course.

[0049] The chlorine necessary for the decomposition can be generated from all of these types of water, and any of these and the treatment target gas may be mixed, followed by irradiation by light so as to be used in the present invention which decomposes the treatment target pollutants.

(Light Irradiation Means)

[0050] As a light irradiation means usable in the present invention, light of, e.g., from 300 to 500 nm wavelength is preferred, and the use of light of from 350 to 450 nm wavelength is more preferred. Also, as light irradiation intensity for the functional water and treatment target, in the case of, e.g., a light source having a peak around 360 nm wavelength, decomposition sufficient in practical use proceeds at an intensity of hundreds of $\mu\text{W}/\text{cm}^2$ (measured between 300 nm and 400 nm). Stated specifically, the irradiation may be made in an amount of light of from 10 $\mu\text{W}/\text{cm}^2$ to 10 mW/cm^2 , and preferably from 50 $\mu\text{W}/\text{cm}^2$ to 5 mW/cm^2 .

[0051] Then, as a light source of such light, natural light (e.g., sunlight) or artificial light (e.g., a mercury lamp, a black light and a color fluorescent lamp) may be used.

[0052] In the present invention, it is unnecessary to use ultraviolet light of about 250 nm or shorter wavelength. Hence, it is also neither necessary to provide any safety device so that human bodies are not affected, nor necessary to make up the decomposition treatment tank using quartz glass through which the ultraviolet light can readily pass. Thus, the system can be set up at a low cost.

(Means for Generating Air Which Contains Chlorine)

[0053] As a means for generating the air which contains chlorine, any device may be used which brings the functional water and the air into contact with each other, e.g., which sends the air to the surface of the functional water. In order to improve efficiency, it is more advantageous to use a device which can ensure a large gas-

liquid contact area. As a means for ensuring such a large contact area, preferred are a means for jetting the functional water in the air in the form of droplets and a means for aerating the functional water.

[0054] These devices may be made of any materials as long as they are not corroded by the treatment target and chlorine. For example, usable are a porous diffusion plate made of sintered glass, porous ceramic, sintered SUS316 stainless steel or a net woven with fibrous SUS316 stainless steel, and a sparger nozzle shower head made of pipes of glass, ceramic or SUS316 stainless steel.

EXAMPLES

[0055] The present invention is described below in greater detail by giving Examples. These Examples by no means limit the present invention.

(Example 1)

[0056] Batch operation of single-unit type decomposition treatment tank, without diaphragm:

[0057] The same decomposition system as the system shown in Fig. 1 but having removed the storage tank 110 therefrom was made ready for use. The electrolytic cell 102 was so set up as to be able to electrolyze about 50 ml of water through a platinum electrode.

[0058] First, the functional water (I) was prepared in the following way using the electrolytic cell 102.

[0059] The electrolyte concentration of water containing sodium chloride as an electrolyte, the electrolysis electric-current value, the electrolysis time and so forth were changed in variety, and the pH of the resultant acidic functional water obtained on the anode side was measured with a pH meter (TCX-90i). The concentration of dissolved chlorine was also measured with a simplified reflection photometer (trade name: RQ flex; manufactured by Merck & Co., Inc.; test paper: Reflectoquant chlorine test paper).

[0060] As a result of this measurement, it was ascertained that the pH of this functional water changed from 4.0 to 10.0 and the concentration of dissolved chlorine from 2 mg/L to 70 mg/L, depending on the concentration of sodium chloride (standard concentration: 1,000 mg/L), the electrolysis electric-current value, the electrolysis time and so forth.

[0061] Accordingly, as the functional water (I) used in the present Example, functional water having a pH of 7.9 and having the dissolved chlorine in a concentration of 15 mg/L was used. This functional water (I) was water obtained by putting 50 mL of distilled water in the electrolytic cell 102, and adding thereto from the electrolyte solution feed unit 105 2 mL of an aqueous sodium chloride solution having a concentration of 20% (250 g/L), to form an aqueous solution of about 1,000 mg/L of sodium chloride, followed by electrolysis for 12 minutes.

[0062] Next, 50 mL of the functional water (I) was put

into a 500 mL volume decomposition treatment tank 101 made of glass.

[0063] In an experiment made previously, this functional water (I) was put into the decomposition treatment tank 101 shown in Fig. 1 and air was sent to the aeration means 107 at a flow rate of 300 mL/min. by means of an air pump. Here, the concentration of chlorine in the gaseous phase portion in the decomposition treatment tank 101 was measured with a detecting tube (manufactured by GASTEC CORPORATION K.K., No. 8H) several times. As a result, it was in the range of from 80 ppm to 300 ppm, but decreased on gradually.

[0064] The gaseous phase portion of this decomposition treatment tank 101 was irradiated by light by means of a black light fluorescent lamp (trade name: FL10BLB; manufactured by Toshiba Corporation; 10 W) which is the light irradiation means 106. This irradiation was made in an amount of light of from 0.4 to 0.7 mW/cm².

[0065] Simultaneously with the irradiation by light, air containing TCE and PCE in a concentration of 100 ppm imitated as polluted air vacuum-extracted from polluted soil, formed using a permeator (manufactured by GASTEC CORPORATION K.K.) was sent at a flow rate of 300 mL/min. from the aeration means 107 provided at the bottom of the decomposition treatment tank 101.

[0066] For 30 minutes after this system was begun to be operated, the concentration of TCE and PCE in the air exhausted from the decomposition treatment tank 101 was periodically checked by sampling using a gas-tight syringe, and the concentration of TCE and PCE was measured by gas chromatography (using GC-14B, trade name; manufactured by Shimadzu Corporation and having an FID detector; column: DB-624, available from J & W K.K.). However, none of them were always detectable. The concentration of TCE and PCE in the functional water was also measured in the same way after the treatment was completed, but none of them were detectable. This showed that the TCE and PCE were decomposable.

[0067] Next, the functional water waste liquor at the bottom of the decomposition treatment tank 101 was all drawn out and was flowed back to the electrolytic cell 102 through the functional water waste liquor flow-back pipe 109 to effect electrolysis again for 12 minutes. As a result, functional water (II) having a pH of 2.3 and having the dissolved chlorine in a concentration of 27 mg/L was formed.

[0068] This functional water (II) was poured into the decomposition treatment tank 101, where the black light fluorescent lamp was again put on and simultaneously aerated with the air containing TCE and PCE.

[0069] In this treatment, too, the concentration of chlorine in the gaseous phase portion in the decomposition treatment tank 101 and the concentration of TCE and PCE in the exhaust air were periodically measured, but none of them were always detectable.

[0070] This operation was carried out five times and

more, but the TCE and PCE came to be included in the exhaust air on the 6th operation and following. Accordingly, the decomposition was once stopped, and the functional water was fed back to the decomposition treatment tank 101. Then, after 2 mL of an aqueous sodium chloride solution was added from the electrolyte solution feed unit 105, the electrolysis was again carried out for 12 minutes, and thereafter the functional water formed was anew supplied to the decomposition treatment tank 101, followed by aeration under irradiation by light from the lamp. As a result, the TCE and PCE became not detectable.

[0071] From this fact, it has been ascertained that the electrolyte may be added at intervals of once in five times when the functional-water waste liquor is batch-wise flowed back, whereby the TCE and PCE can be continued being decomposed while the functional-water waste liquor having been aerated is electrolyzed and re-generated into the functional water (II) which is again utilized as the feed source of the air which contains chlorine.

(Example 2)

[0072] Continuous operation of single-unit type decomposition treatment tank, without diaphragm:

[0073] The same decomposition system as the system shown in Fig. 1 but having removed the storage tank 110 therefrom was made ready for use.

[0074] 50 mL of functional water (I) formed in the same manner as in Example 1 was put into the decomposition treatment tank 101. Subsequently, 50 mL of an aqueous sodium chloride solution having a concentration of 1,000 mg/L was put into the electrolytic cell 102 to effect electrolysis, during which the functional water was supplied from the electrolytic cell 102 at a flow rate of 2 mL/min. by means of a pump. Also, the functional water waste liquor having been aerated was drained off at the same rate so as to be flowed back by 100% to the electrolytic cell 102 and so that the functional water in the decomposition treatment tank 101 and electrolytic cell 102 was kept in a constant quantity.

[0075] In an experiment made previously, air was sent to the aeration means 107 at a flow rate of 300 mL/min. by means of an air pump while the functional water was circulated between the decomposition treatment tank 101 and the electrolytic cell 102. Here, the concentration of chlorine in the gaseous phase portion in the decomposition treatment tank 101 was measured with a detecting tube (manufactured by GASTEC CORPORATION K.K., No. 8H) several times. As a result, it was in the range of from 80 ppm to 300 ppm in the beginning, but increased on gradually.

[0076] This decomposition treatment tank 101 was irradiated by light of a black light fluorescent lamp and simultaneously the air containing TCE and PCE in a concentration of 100 ppm was sent at a flow rate of 300 mL/min., in the same manner as in Example 1.

[0077] For about 4 hours after this system was begun to be operated, the concentration of TCE and PCE in the air exhausted from the decomposition treatment tank 101 was periodically checked by sampling using a gas-tight syringe, and the concentration of TCE and PCE was measured in the same manner as in Example 1. However, none of them were always detectable. This showed that the TCE and PCE were decomposable only by again electrolyzing the functional water waste liquor into the functional water (II) followed by aeration, in the course where the functional water is circulated five times in through the system.

[0078] After that, however, the TCE and PCE came to be included in the exhaust air. Accordingly, 4 mL of an aqueous sodium chloride solution was added to the electrolytic cell 102 from the electrolyte solution feed unit 105 little by little over a period of about 30 minutes. As a result, the TCE and PCE became not detectable.

[0079] From this fact, it has been ascertained that the electrolyte may be added every time the water in the system is circulated five times when the functional water waste liquor is continuously flowed back, whereby the TCE and PCE can be continued being decomposed while the functional water waste liquor having been aerated is electrolyzed and regenerated into the functional water (II) which is again utilized as the feed source of the air which contains chlorine.

(Example 3)

[0080] Batch operation of separation type decomposition treatment tank, without diaphragm:

[0081] Using the same decomposition system as the system shown in Fig. 1 except that the functional water aeration tank 201 was separate from the decomposition treatment tank 101, an experiment was made in the same manner as in Example 1. Here, the functional water aeration tank 201 was 70 mL in volume, and 50 mL of functional water was put into it. Also, the decomposition treatment tank 101 was 450 mL in volume.

[0082] As a result, entirely the same results as those in Example 1 were obtained.

[0083] From this fact, it has been ascertained that, also where the functional water aeration tank 201 is separate from the decomposition treatment tank 101, the electrolyte may be added every time the water in the system is circulated five times when the functional-water waste liquor is batch-wise flowed back, whereby the TCE and PCE can be continued being decomposed while the functional water waste liquor having been aerated is electrolyzed and regenerated into the functional water (II) which is again utilized as the feed source of the air which contains chlorine.

(Example 4)

[0084] Batch operation of aeration type decomposition treatment tank using air not containing any pollut-

ants, without diaphragm:

[0085] Using the same decomposition system as the system shown in Fig. 1 except that the polluted air from the permeator was directly sent to the decomposition treatment tank 101 and 300 mL/min. of the air not containing any pollutants was sent to the aeration means 107 provided at the bottom of the decomposition treatment tank 101, at a flow rate of 300 mL/min. by means of an air pump, an experiment was made in the same manner as in Example 1.

[0086] As a result, entirely the same results as those in Example 1 were obtained.

[0087] From this fact, it has been ascertained that, also where the functional water is aerated with the air not containing any pollutants to form the air which contains chlorine which is then mixed with pollutants in the decomposition treatment tank 101, the electrolyte may be added every time the water in the system is circulated five times when the functional-water waste liquor is batch-wise flowed back, whereby the TCE and PCE can be continued being decomposed while the functional water waste liquor having been aerated is electrolyzed and regenerated into the functional water (II) which is again utilized as the feed source of the air which contains chlorine.

(Example 5)

[0088] Batch operation of single-unit type decomposition treatment tank, with diaphragm:

[0089] An experiment was made using a system in which as shown in Fig. 3 the diaphragm 111 was attached to the electrolytic cell 102 and the alkaline-water pipe 112 was provided on the cathode side.

[0090] In the same manner as in Example 1, the electrolyte concentration of water containing sodium chloride as an electrolyte, the electrolysis electric-current value, the electrolysis time and so forth were changed in variety, and the pH and concentration of dissolved chlorine of the resultant acidic functional water obtained on the anode side were measured with a pH meter (TCX-90i).

[0091] As a result of this measurement, it was ascertained that the pH of this functional water changed from 1.0 to 4.0 and the concentration of dissolved chlorine from 5 mg/L to 150 mg/L, depending on the concentration of sodium chloride (standard concentration: 1,000 mg/L), the electrolysis electric-current value, the electrolysis time and so forth.

[0092] Accordingly, as the functional water (I) used in the present Example, functional water having a pH of 2.1 and having the dissolved chlorine in a concentration of 60 mg/L was used. This functional water (I) was 50 mL of acidic electrolytic water obtained on the side of the anode 104 by putting 100 mL of distilled water in the electrolytic cell 102, and adding thereto from the electrolyte solution feed unit 105 4 mL of an aqueous sodium chloride solution having a concentration of 20% (250 g/

L), to form an aqueous solution of about 1,000 mL of sodium chloride, followed by electrolysis for 12 minutes.

[0093] This functional water (I) was supplied to the decomposition treatment tank 101 in the same manner as in Example 1, and an experiment was made in the same manner as in Example 1 except that the concentration of the air containing TCE and PCE was doubled to 200 ppm. With regard to the functional water waste liquor, it was neutralized in the functional water waste liquor flow-back pipe 109 by supplying from the alkaline-water pipe 112 50 mL of alkaline water formed on the side of the cathode of the electrolytic cell 102, thereafter temporarily stored in the storage tank 110 and then flowed back again to the electrolytic cell 102. Here, the pH of the functional water waste liquor having not been neutralized was 2.3. After neutralization, it was 6.8.

[0094] As a result, entirely the same results as those in Example 1 were obtained.

[0095] From this fact, it has been ascertained that, where the functional water formed in the electrolytic cell having a diaphragm is used, the electrolyte may be added every time the water in the system is circulated five times when the functional-water waste liquor is batch-wise flowed back, whereby the TCE and PCE can be continued being decomposed while the functional water waste liquor having been aerated is electrolyzed and regenerated into the functional water (II) which is again utilized as the feed source of the air which contains chlorine.

(Example 6)

[0096] Batch operation of single-unit type decomposition treatment tank, using functional water with hypochlorite:

[0097] An experiment was made in the same manner as in Example 5 except that 50 mL of functional water (I) made up by adding hydrochloric acid, sodium chloride and sodium hypochlorite was put into the decomposition treatment tank 101 at the time the experiment was started.

[0098] The functional water (I) was prepared by adding to distilled water the hydrochloric acid, sodium chloride and sodium hypochlorite so as to be in concentrations of 0.006 mol/L, 0.01 mol/L and 0.002 mol/L, respectively. Here, the functional water (I) had a pH of 2.3 and had the dissolved chlorine in a concentration of 110 mg/L.

[0099] This functional water (I) was supplied to the decomposition treatment tank 101 in the same manner as in Example 5, and an experiment was made in the same manner as in Example 5 except that, to the functional-water waste liquor having been flowed back, sodium chloride was added from the electrolyte solution feed unit 105 so as to be in a concentration of 1,000 mg/L and thereafter the electrolysis was carried out.

[0100] As a result, quite the same results as those in Example 1 were obtained, except that the TCE and PCE

came to be included in the exhaust air when the functional water (II) formed after the functional water waste liquor was flowed back seven times to effect electrolysis repeatedly was used.

[0101] From this fact, it has been ascertained that, where the functional water with hypochlorite is used, the electrolyte may be added every time the water in the system is electrolyzed six times to regenerate the functional water (II), whereby the TCE and PCE can be continued being decomposed while the functional water waste liquor having been aerated is electrolyzed and regenerated into the functional water (II) which is again utilized as the feed source of the air which contains chlorine.

Claims

1. A pollutant decomposition process for decomposing pollutants by bringing pollutants contained in air into contact with air which contains chlorine, under irradiation by light, the process comprising:

a chlorine-containing air generation step of generating air which contains chlorine, by bringing air into contact with a chlorine-generating solution comprised of functional water (I) or functional water (II) having been fed into a chlorine generation region;

a decomposition step of decomposing the pollutants by bringing the air which contains chlorine and air which contains pollutants into contact with each other under irradiation by light in a decomposition treatment region;

a regeneration step of obtaining functional water (II) by regeneration by feeding as functional water waste liquor at least part of the chlorine-generating solution present in the chlorine generation region, to means for forming functional water by electrolysis; and

a feed step of feeding to the chlorine generation region the functional water (II) obtained through the regeneration step;

the functional water (I) and functional water (II) being water capable of generating by aeration the air which contains chlorine, and the functional water (I) comprising a solution used for its formation which does not contain the functional water waste liquor and the functional water (II) comprising a solution used for its formation which contains the functional water waste liquor at least in part.

2. The pollutant decomposition process according to claim 1, wherein the step of electrolyzing the functional water waste liquor to form the functional water (II) is repeated a plurality of times.

3. The pollutant decomposition process according to claim 2, wherein an electrolyte is added to the functional water waste liquor at intervals of once in five or less times when the functional water waste liquor is electrolyzed.
4. The pollutant decomposition process according to claim 1, wherein in the chlorine-containing air generation step the air is brought into contact with the functional water (I) and functional water (II) through the step of sending air to the surfaces of the functional water (I) and functional water (II), the step of jetting the functional water (I) and functional water (II) into air, or the step of aerating the functional water (I) and functional water (II).
5. The pollutant decomposition process according to claim 4, wherein the chlorine generation region constitutes a decomposition treatment tank having the decomposition treatment region in a single unit, and lies at the lower part thereof.
6. The pollutant decomposition process according to claim 4, wherein the chlorine generation region comprises a functional water aeration tank provided separately, and only the gas generated is sent to the decomposition treatment tank consisting only of the decomposition treatment region.
7. The pollutant decomposition process according to claim 1, wherein the means for forming functional water by electrolysis does not have any diaphragm.
8. The pollutant decomposition process according to claim 1, wherein the functional water waste liquor is neutralized before the functional water waste liquor is electrolyzed.
9. The pollutant decomposition process according to claim 8, wherein alkaline water formed by electrolysis of a solution containing an electrolyte is used to neutralize the functional water waste liquor.
10. The pollutant decomposition process according to claim 1, wherein at least one of the functional water (I) and the functional water (II) is acidic water formed by electrolysis of a solution containing an electrolyte.
11. The pollutant decomposition process according to any one of claims 3, 9 and 10, wherein the electrolyte is at least one of sodium chloride and potassium chloride.
12. The pollutant decomposition process according to claim 1, wherein the functional water (I) is a functional water containing a hypochlorite or an aqueous hypochlorite solution.
13. The pollutant decomposition process according to claim 12, wherein the hypochlorite is at least one of sodium hypochlorite and potassium hypochlorite.
14. The pollutant decomposition process according to claim 12 or 13, wherein the functional water (I) contains an inorganic acid or an organic acid.
15. The pollutant decomposition process according to claim 14, wherein the inorganic acid or organic acid is at least one acid selected from hydrochloric acid, hydrofluoric acid, oxalic acid, sulfuric acid, phosphoric acid, boric acid, acetic acid, formic acid, malic acid and citric acid.
16. The pollutant decomposition process according to claim 1, wherein the functional water (I) and functional water (II) have a hydrogen ion concentration (pH value) of from 1 to 4 and a dissolved-chlorine concentration of from 5 mg/L to 150 mg/L.
17. The pollutant decomposition process according to claim 1, wherein the functional water (I) and functional water (II) have a hydrogen ion concentration (pH value) of from 4 to 10 and a dissolved-chlorine concentration of from 2 mg/L to 100 mg/L.
18. The pollutant decomposition process according to claim 1, wherein the light for irradiation comprises light having a wavelength in the wave range of from 300 nm to 500 nm.
19. The pollutant decomposition process according to claim 1, wherein the light for irradiation comprises light having a wavelength in the wave range of from 350 nm to 450 nm.
20. The pollutant decomposition process according to claim 1, wherein the irradiation is made in an amount of light of from 10 $\mu\text{W}/\text{cm}^2$ to 10 mW/cm^2 .
21. The pollutant decomposition process according to claim 1, wherein the irradiation is made in an amount of light of from 50 $\mu\text{W}/\text{cm}^2$ to 5 mW/cm^2 .
22. The pollutant decomposition process according to claim 1, wherein the air which contains pollutants is air vacuum-extracted from soil which contains pollutants.
23. The pollutant decomposition process according to claim 1, wherein the air which contains pollutants is air formed by aeration of underground water which contains pollutants.
24. The pollutant decomposition process according to claim 1, wherein the pollutants comprise an organochlorine compound.

25. The pollutant decomposition process according to claim 24, wherein the organochlorine compound is a compound selected from the group consisting of chloroethylene, 1,1-dichloroethylene, cis-1,2-dichloroethylene, trans-1,2-dichloroethylene, trichloroethylene, tetrachloroethylene, chloromethane, dichloromethane and trichloromethane.
26. A pollutant decomposition system for decomposing pollutants by bringing pollutants contained in air into contact with air which contains chlorine, under irradiation by light, the system comprising:
- a chlorine generation region into which a chlorine-generating solution comprising functional water (I) or functional water (II) is fed to bring it into contact with air to generate air which contains chlorine;
 - a decomposition treatment region into which the air which contains chlorine and air which contains pollutants are fed to bring them into contact with each other under irradiation by light;
 - means for effecting irradiation by light;
 - means for forming functional water by electrolysis; and
 - means for feeding at least part of the chlorine-generating solution to the means for forming functional water;
 - the functional water (I) and functional water (II) being water capable of generating by aeration the air which contains chlorine, and the functional water (I) comprising a solution used for its formation which does not contain the functional water waste liquor and the functional water (II) comprising a solution used for its formation which contains the functional water waste liquor at least in part.
27. The pollutant decomposition system according to claim 26, wherein in the chlorine generation region the air is brought into contact with the functional water (I) and functional water (II) through means for sending air to the surfaces of the functional water (I) and functional water (II), means for jetting the functional water (I) and functional water (II) into air, or means for aerating the functional water (I) and functional water (II).
28. The pollutant decomposition system according to claim 27, wherein the chlorine generation region constitutes a decomposition treatment tank having the decomposition treatment region in a single unit, and lies at the lower part thereof.
29. The pollutant decomposition system according to claim 27, which comprises a functional water aeration tank provided separately to constitute the chlorine generation region, and a decomposition treatment tank consisting only of the decomposition treatment region.
30. The pollutant decomposition system according to claim 26, wherein the means for forming functional water by electrolysis does not have any diaphragm.
31. The pollutant decomposition system according to claim 26, wherein the functional water waste liquor is neutralized before the functional water waste liquor is electrolyzed.
32. The pollutant decomposition system according to claim 31, wherein alkaline water formed by electrolysis of a solution containing an electrolyte is used to neutralize the functional water waste liquor.
33. The pollutant decomposition system according to claim 26, wherein at least one of the functional water (I) and the functional water (II) is acidic water formed by electrolysis of a solution containing an electrolyte.
34. The pollutant decomposition system according to claim 32 or 33, wherein the electrolyte is at least one of sodium chloride and potassium chloride.
35. The pollutant decomposition system according to claim 26, wherein the functional water (I) is a functional water containing a hypochlorite or an aqueous hypochlorite solution.
36. The pollutant decomposition system according to claim 35, wherein the hypochlorite is at least one of sodium hypochlorite and potassium hypochlorite.
37. The pollutant decomposition system according to claim 35 or 36, wherein the functional water (I) contains an inorganic acid or an organic acid.
38. The pollutant decomposition system according to claim 37, wherein the inorganic acid or organic acid is at least one acid selected from hydrochloric acid, hydrofluoric acid, oxalic acid, sulfuric acid, phosphoric acid, boric acid, acetic acid, formic acid, malic acid and citric acid.
39. The pollutant decomposition system according to claim 26, wherein the functional water (I) and functional water (II) have a hydrogen ion concentration (pH value) of from 1 to 4 and a dissolved-chlorine concentration of from 5 mg/L to 150 mg/L.
40. The pollutant decomposition system according to claim 26, wherein the functional water (I) and functional water (II) have a hydrogen ion concentration (pH value) of from 4 to 10 and a dissolved-chlorine

concentration of from 2 mg/L to 100 mg/L.

41. The pollutant decomposition system according to claim 26, wherein the light for irradiation comprises light having a wavelength in the wave range of from 300 nm to 500 nm. 5
42. The pollutant decomposition system according to claim 26, wherein the light for irradiation comprises light having a wavelength in the wave range of from 350 nm to 450 nm. 10
43. The pollutant decomposition system according to claim 26, wherein the irradiation is made in an amount of light of from 10 $\mu\text{W}/\text{cm}^2$ to 10 mW/cm^2 . 15
44. The pollutant decomposition system according to claim 26, wherein the irradiation is made in an amount of light of from 50 $\mu\text{W}/\text{cm}^2$ to 5 mW/cm^2 . 20
45. The pollutant decomposition system according to claim 26, wherein the air which contains pollutants is air vacuum-extracted from soil which contains pollutants. 25
46. The pollutant decomposition system according to claim 26, wherein the air which contains pollutants is air formed by aeration of underground water which contains pollutants. 30
47. The pollutant decomposition system according to claim 26, wherein the pollutants comprise an organochlorine compound. 35
48. The pollutant decomposition system according to claim 47, wherein the organochlorine compound is a compound selected from the group consisting of chloroethylene, 1,1-dichloroethylene, cis-1,2-dichloroethylene, trans-1,2-dichloroethylene, trichloroethylene, tetrachloroethylene, chloromethane, dichloromethane and trichloromethane. 40

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FIG. 1

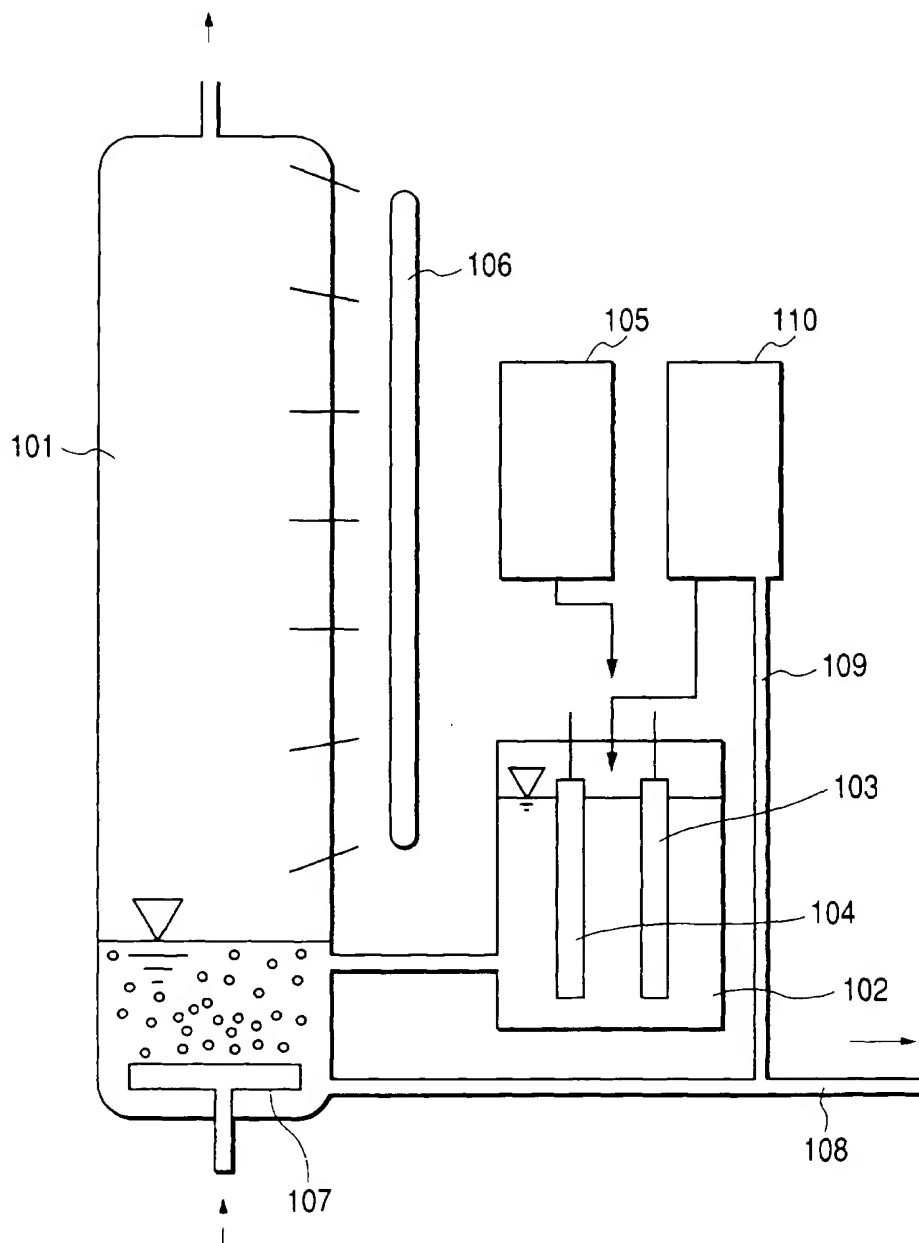


FIG. 2

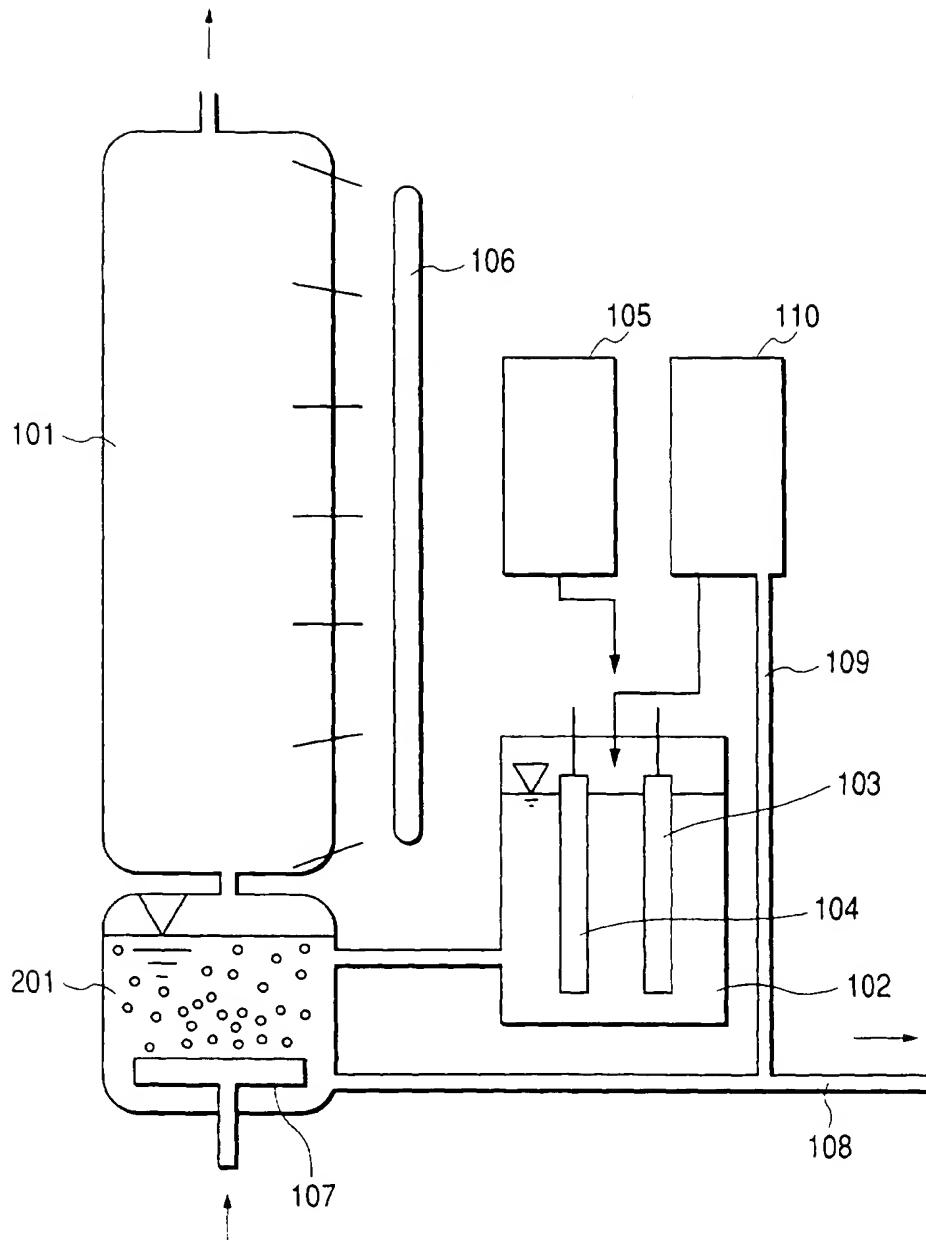
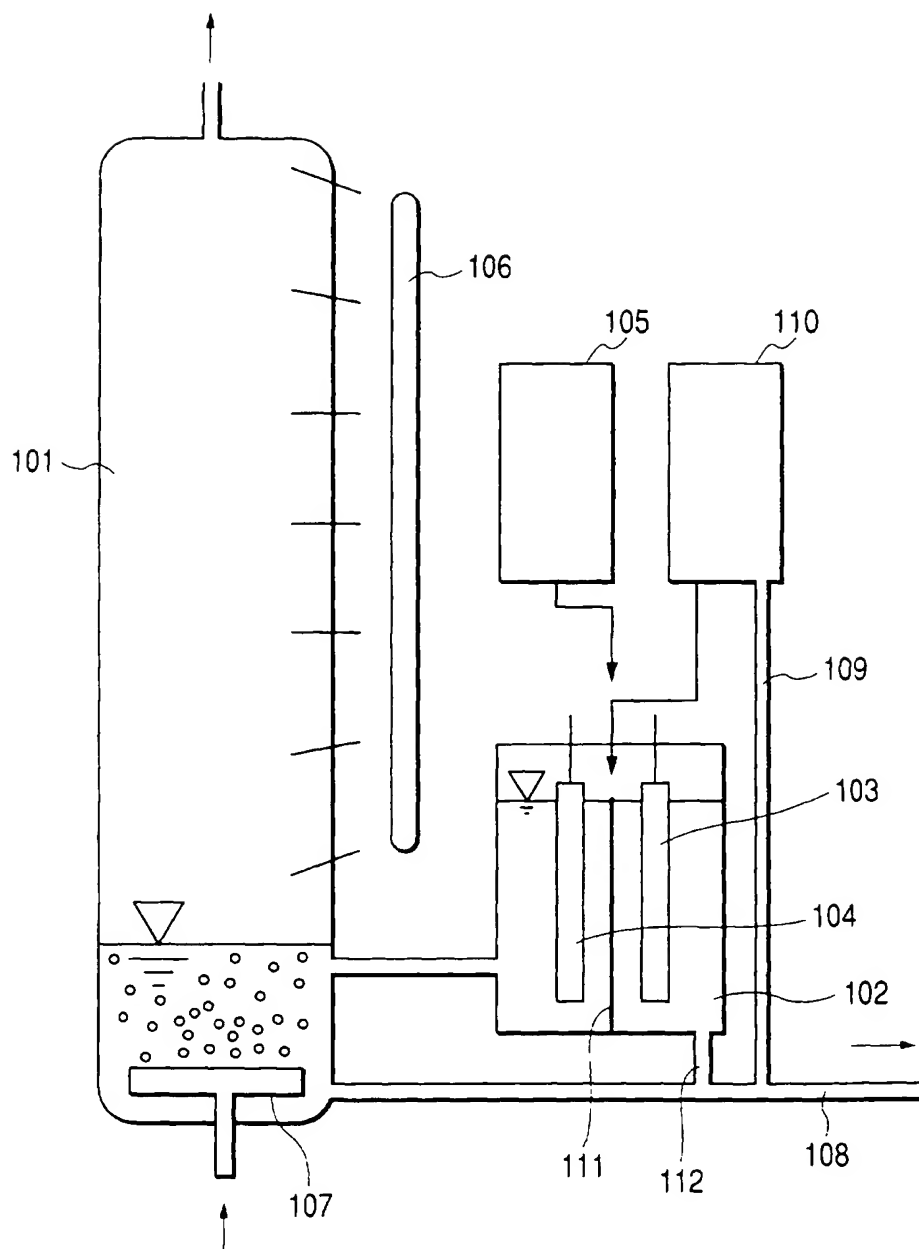


FIG. 3





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EUROPEAN SEARCH REPORT

Application Number
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